NOLTR 71-103

GAMMA-RAY SPECTRA OF FRACTIONATED FISSION PRODUCTS

By
L. R. Bunney
D. Sam

18 JUNE 1971

NOL

NAVAL ORDNANCE LABORATORY, WHITE OAK, SILVER SPRING, MARYLAND

NOLTR 71-103

UNCLASSIFIED
Security Classification

DOCUMENT CONTROL DATA - R & D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY (Corporate author)	2a. REPORT SECURITY CLASSIFICATION
Naval Ordnance Laboratory Silver Spring, Maryland 20910	Unclassified
	2b. GROUP

3. REPORT TITLE

GAMMA-RAY SPECTRA OF FRACTIONATED FISSION PRODUCTS

4. DESCRIPTIVE NOTES (Type of report and inclusive dates)

Final Report

5. AUTHOR(S) (First name, middle initial, last name)

Bunney, Leland R.
Sam, Daniel

6. REPORT DATE	7a. TOTAL NO. OF PAGES	7b. NO. OF REFS
18 June 1971	35	21

8a. CONTRACT OR GRANT NO.	9a. ORIGINATOR'S REPORT NUMBER(S)
Work Order No. DAHC20-70-C-0283	NOLTR 71-103
b. PROJECT NO. OCD Work Unit 3125A	
c.	9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report)
d.	

10. DISTRIBUTION STATEMENT

Approved for public release; distribution unlimited

11. SUPPLEMENTARY NOTES	12. SPONSORING MILITARY ACTIVITY
	Office of Civil Defense Office of the Secretary of the Army Washington, D. C. 20310

13. ABSTRACT

To determine the effects of fractionation on gamma-ray exposure rates in fission-product fields, spectra of gamma-rays emitted by fractionated products of thermal neutron fission of ^{235}U were studied. Controlled fractionation was brought about by sweeping the rare gas fission products out of sealed samples at 10 to 15 seconds after fission using programmed automatic equipment. Spectra were measured at nine selected times (viz., 1/4, 1/2, 1, 2, 5, 10, 24, 48 and 72 hours) after fission. The detector used was a calibrated and highly collimated 5" x 5" NaI(Tl) crystal. The gamma-ray spectra were unfolded from the pulse-height distributions by means of an iterative method. The number of fissions that had occurred in each sample was determined by radiochemical analysis for ^{99}Mo.

The type and time of fractionation studied in this work is of particular interest in predicting the effects of nuclear explosions, but is also applicable to certain possible nuclear reactor incidents.

14 KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Fission products Gamma-ray spectra Fractionation Exposure rates Dose rates Scintillation spectrometry						

NOLTR 71-103

GAMMA-RAY SPECTRA OF FRACTIONATED FISSION PRODUCTS

by
L. R. Bunney
D. Sam

for

Office of Civil Defense
Office of the Secretary of the Army
Washington, D. C. 20310

Final Report
Work Order No. DAHC20-70-C-0283
(OCD Work Unit 3125A)

OCD Review Notice:

This report has been reviewed in the Office of Civil Defense and approved for publication. Approval does not signify that the contents necessarily reflect the views and policies of the Office of Civil Defense.

Approved for Public Release
Distribution Unlimited

GAMMA-RAY SPECTRA OF FRACTIONATED FISSION PRODUCTS

by
L. R. Bunney
D. Sam

ABSTRACT: To determine the effects of fractionation on gamma-ray exposure rates in fission-product fields, spectra of gamma-rays emitted by fractionated products of thermal neutron fission of ^{235}U were studied. Controlled fractionation was brought about by sweeping the rare gas fission products out of sealed samples at 10 to 15 seconds after fission using programmed automatic equipment. Spectra were measured at nine selected times (viz., 1/4, 1/2, 1, 2, 5, 10, 24, 48 and 72 hours) after fission. The detector used was a calibrated and highly collimated 5" x 5" NaI(Tl) crystal. The gamma-ray spectra were unfolded from the pulse-height distributions by means of an iterative method. The number of fissions that had occurred in each sample was determined by radiochemical analysis for ^{99}Mo.

The type and time of fractionation studied in this work is of particular interest in predicting the effects of nuclear explosions, but is also applicable to certain possible nuclear reactor incidents.

NUCLEAR CHEMISTRY DIVISION
CHEMISTRY RESEARCH DEPARTMENT
NAVAL ORDNANCE LABORATORY
WHITE OAK, MARYLAND

NOLTR 71-103 18 June 1971

Gamma-Ray Spectra of Fractionated Fission Products

The work reported herein was supported by the Office of Civil Defense, Work Order No. DAHC20-70-C-0283 (Work Unit 3125A).

The authors would like to express their gratitude for the encouragement of Dr. N. E. Ballou of Battelle Northwest Laboratories, under whose direction this work was begun. We are very appreciative of the interest, understanding and continuing support of Mr. J. C. Green and Dr. D. W. Bensen of the Office of Civil Defense. We wish to thank Mr. R. L. Tomlinson and the staff of the Aerojet-General reactor facility for their exceptional cooperation and many kindnesses.

GEORGE G. BALL
Captain, USN
Commander

ALBERT LIGHTBODY
By direction

CONTENTS

ILLUSTRATIONS

TABLES

INTRODUCTION

In studies of nuclear bomb debris, fractionation of the radioactive species resulting from the detonation has been observed on many occasions (1,2,3). The phenomenon of fractionation is defined by Freiling (4) as "any alteration in composition which caused the debris sample to be nonrepresentative of the detonation products taken as a whole". Fractionation can be caused by a number of different processes and can occur at various times after detonation. Freiling has discussed fractionation in several papers (4,5,6). The work of Adams et al (7,8) and that of Norman et al (9,10) is of interest in consideration of fractionation processes. Many others have made contributions to fractionation information; among them are those who have participated in symposia (11,12) on fallout.

According to some calculations (6,13), it is possible for fractionation to have a large effect on the gamma-rays emitted by the fission products in fallout. To our knowledge, no laboratory studies have been performed on the radiological effects of fractionation under controlled conditions.

In previous work (14-18) we have measured the gamma-rays from unfractionated products of fission as a function of time. The products studied were those of slow neutron fission of ^{235}U and ^{238}U. The work described here was undertaken to show quantitatively the effect of very early rare gas fractionation. This is only one of many possible fractionation processes, but it can result in very marked losses or enhancements of certain mass chains in nuclear debris.

The studies involved the measurement of the gamma-ray pulse-height distributions from fission products that had been fractionated in the laboratory in a controlled manner. Pulse-height distributions were taken on the fraction of the fission products remaining after removing the rare gases at the same times after fission as in the studies of unfractionated fission products (viz., 1/4, 1/2, 1, 2, 5, 10, 24, 48 and 72 hours). In addition, pulse-height distributions were obtained at 1 and 2 hours after fission on the decay products of the rare gases which were removed at 10-15 seconds after the midpoint of a 10 second irradiation. The time at which the rare gases were removed (viz., 10-15 seconds after fission), corresponds approximately to the condensation time of a nuclear weapon of about 10 KT yield.

The results of the work show the nature and extent of the decrease in photon emission and exposure rates due to the early removal of the rare gas fission products and the consequent lack of

descendants of these products at later times in the main body of the fision product mixture.

EXPERIMENTAL

In order to measure the desired gamma-ray spectra, gamma-ray pulse-height distributions were obtained on the fractions of gross fission products remaining after the removal of the rare gases. The same nine elapsed times selected for the unfractionated work were used in order to permit a direct comparison of the fractionation effects. In addition to the measurements on the solid residue, the rare gases were trapped and the gamma-rays of their descendants were examined at the widely used one-hour-post-fission point, and also at two hours after fission.

The main steps in the experimental procedure were: (1) assembly of the detector shield and pulse-height analyzer system, (2) calibration of the detector, (3) preparation and packaging of the samples, (4) irradiation of the samples, (5) collection of the rare gas fission products, (6) measurement of the pulse-height distributions, (7) readout of the data in a form suitable for machine computation, and (8) radiochemical determination of the number of fission events in each sample. Steps (1), (2), (6), (7) and (8) were performed in a manner that was essentially identical with previous descriptions (14,17).

Preparation and Packaging of Samples

Uranyl stearate was prepared by mixing warm aqueous solutions of sodium stearate and uranyl nitrate in a similar fashion to that used by Wahl (19) in preparing barium stearate. First the enriched uranyl nitrate was obtained by dissolving uranium metal foil in dilute HNO_3, evaporating to dryness and dissolving in distilled H_2O. The original foil had an abundance of 93.2% of the mass-235 isotope. Spectrographic analysis had shown that the foil was 99.8% pure uranium. Uranyl stearate was used because it was reported to be highly effective in emanating rare gas fission products. Its emanating power was determined by Wahl (20) to be >95% for the 3-sec ^{92}Kr. The use of enriched uranium permitted the physical dimensions of the sample to be small enough to make good counting sources.

Packaging for irradiation and counting was designed to prevent the escape of any fission fragments from the samples, while permitting subsequent radiochemical determination of ^{99}Mo. It was also necessary to design the packaging for the rapid removal of the gaseous fission products from the uranyl stearate samples at a desired time after fission. Therefore, the uranyl stearate powder was sandwiched between Whatman #42 filter paper discs and then heat-sealed between polyvinyl alcohol films as described below. Polyvinyl alcohol film was selected because it is acid-soluble and may be dissolved with the entire uranyl stearate packet for the subsequent determination of ^{99}Mo. This determination was used to calculate the number of fissions that had occurred in the sample.

A cylindrical cavity having a volume of about 0.75 ml was provided in the sample for holding the uranyl stearate. This cavity was formed by stacking eight pieces of filter paper which had been cut into rings with an inside diameter of 0.80 cm and an outside diameter of 1.43 cm. The stack of rings whose inner core was filled with uranyl stearate, was sandwiched between two filter paper discs of the same outside diameter. Approximately 36 mg. of uranyl stearate was introduced into the cavity for each sample.

This packet of filter papers was pressed tightly together and heat-sealed between two pieces of .08 mm thick polyvinyl alcohol film. It was proposed to pentrate this packet with hollow needles for the purpose of withdrawing the gaseous fission products. In order to provide a leak-proof seal when these needles were withdrawn, a self-sealing rubber gasket was placed on one side of the packet, and another piece of polyvinyl alcohol film was placed on top of the gasket and again heat-sealed. This arrangement gave a very tight seal, and kept the uranyl stearate firmly in place.

The uranyl stearate package was placed in a high density nylon rabbit. Nylon was used because it is tough and can withstand the force of impact after transport in or out of the reactor. The specially designed rabbit (cf Fig. 1) was composed of two parts, which screwed together. The cap was 2.44 cm long with a circular opening for needle penetration into the uranyl stearate sample. The body was a solid piece of nylon 2.44 cm long. A polyethylene disc on top and up to six additional filter paper discs beneath the packet were used to position the uranyl stearate precisely when the cap was screwed tightly onto the body. The overall dimension of the rabbit when assembled was 2.49 cm outside and approximately 3 cm long.

Irradiation and Counting of Samples

The samples were irradiated in a pneumatic rabbit facility of the reactor of the Aerojet-General Corporation at San Ramon, California. This reactor can operate at power levels up to 250 KW. At the maximum power level the slow (<1 ev) neutron flux in the rabbit facility was $\sim 4 \times 10^{12} n/(cm^2\text{-sec})$. The Cd ratio for Au was about 4.5.

In Table I are given the reactor power level and the counting periods (relative to the midpoint of the irradiations) of the pulse-height measurements. The time intervals were measured with a stop watch or electric clock. The duration of each irradiation was 10 sec. Power levels were chosen to provide samples that would give the best possible counting statistics, without producing excessive dead time in the pulse-height analyzer or creating instrumental calibration shifts. In general, the relative dead time for the shorter counting intervals was ~10% and did not exceed 20%. The live-time for each counting interval was obtained from a live timer. The counting intervals were chosen so as to give good counting statistics and to minimize decay effects.

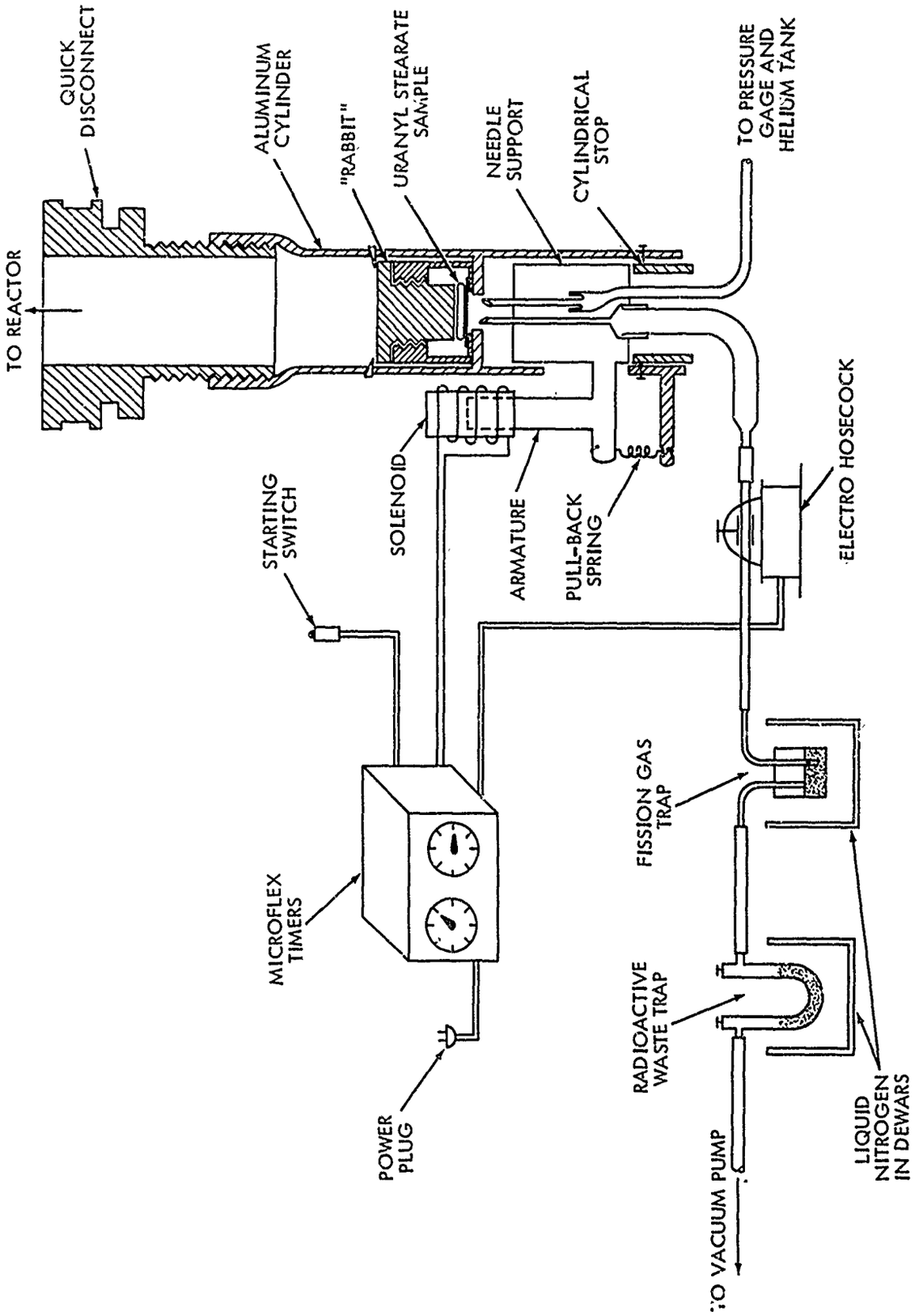

FIG. 1 SEMI-AUTOMATIC FISSION GAS COLLECTION SYSTEM (NOT TO SCALE).

Table 1

Counting Times of Samples

Elapsed time after Mid-Point of Irradiation (hr)	Reactor Power (kw)	Period of count (after Mid-Point of Irradiation)
1/4	40	14-16 min
1/2	40	28-30 min
1	125	57-63 min
2	125	114-126 min
5	250	4.75-5.25 hr
10	250	9.5-10.5 hr
24	250	23-25 hr
48	250	46-50 hr
72	250	69-75 hr

Prior to counting, each uranyl stearate sample was fastened with tape to a thin sheet of polyethylene that was stretched over a polyethylene β-ray absorber 4.3 cm thick. Reproducible geometry was obtained by placement of the sample in the center of a series of concentric rings drawn on the polyethylene absorber. However, for countint of the gaseous fission product samples, a different polyethylene β-ray absorber was used. It had the same overall dimensions, but had a cylindrical well cut into it in order to position the samples for counting. The well was 1.59 cm diameter and 0.343 cm deep. The design of this arrangement was such that the activated charcoal containing the gaseous fission products would have very nearly the same counting geometry as the solid uranyl stearate sample. This made possible the direct comparison of the respective pulse-height data of the solid and the gaseous fission product fractions.

The pulse-height data were recorded and made ready for entry into the computer in the same manner as described in previous reports (14, 17).

Collection of Gaseous Fission Products

A semi-automatic gas-trapping system was constructed. The apparatus was composed of 3 parts: a snap-on rabbit receiver assembly, a gaseous fission product collection train, and a semi-automatic timing circuit. Figure 1 shows schematically how the apparatus was assembled.

The snap-on rabbit receiver assembly was made up of four parts: a quick disconnect, an aluminum cylinder, a movable needle support, and a solenoid.

The brass quick disconnect is commercially available. It was screwed into an aluminum cylinder which contained a top and a bottom compartment separated by a 0.32 cm thick partition. A circular hole, 1.19 cm diameter, was drilled through the center of this partition. The top compartment received the nylon rabbit after irradiation. This compartment was fitted with four spring-loaded pawls which snapped in behind the rabbit on its arrival from the reactor and held it tightly in position. Each pawl was cut to a slightly different length to allow for some variation in rabbit length.

Two 22-gauge stainless steel syringe needles were silver-soldered to a movable brass support which was fitted loosely into the bottom compartment. The two needles were separated by a distance of 0.95 cm. They were positioned in the support so that the needle points would penetrate into the filter paper packet surrounding the irradiated uranyl stearate sample, but would not enter the cavity containing the uranyl stearate. One needle was a gas-entry needle through which helium was introduced for sweeping the gases out of the irradiated uranyl stearate sample. The other needle was a gas-exhaust needle through which the gases from the sample were withdrawn.

A solenoid was clamped firmly to the aluminum cylinder. The armature of the solenoid was bolted to the brass needle support, so that the needles moved up or down as the solenoid was energized or de-energized. The needle support was seated on a cylindrical stop by a pull-back spring when the solenoid was de-energized.

Rubber tubing was used to connect the gas-entry needle to a tank of helium through a flowmeter and a supply valve. The flow-rate used was approximately 75 cc/min to provide a strong flow of helium during the gas-sweeping process. A hypodermic glass syringe (Yale type, 1 cc volume) was pressure fitted into the exhaust needle. Rubber tubing, connected to the syringe, was passed through a Fischer electrohosecock and connected to one arm of a gaseous fission product collection trap cooled in liquid nitrogen. The other arm of this trap was connected to one arm of a larger U-tube trap containing activated charcoal cooled in liquid nitrogen. This U-tube was used to absorb waste gaseous radioactive products to prevent them from escaping into the atmosphere. Care was necessary when handling this trap, because it could become saturated with atmospheric gases which would expand rapidly and dangerously upon warming. The other arm of the trap was connected to a vacuum pump for evacuation of the air to permit a rapid sweeping of the sample with He.

The trap for the collection of gaseous fission products is shown in Fig. 2. It was made from a stainless steel pipe cap and plug with a 1/8 in. pipe thread. The bottom of the cap was machined to 0.010 in. thickness to minimize gamma-ray absorption. Two holes, each about 1/8 in. diameter, were drilled through the plug. Copper tubing 1/8 in. diameter was inserted through each hole and silver

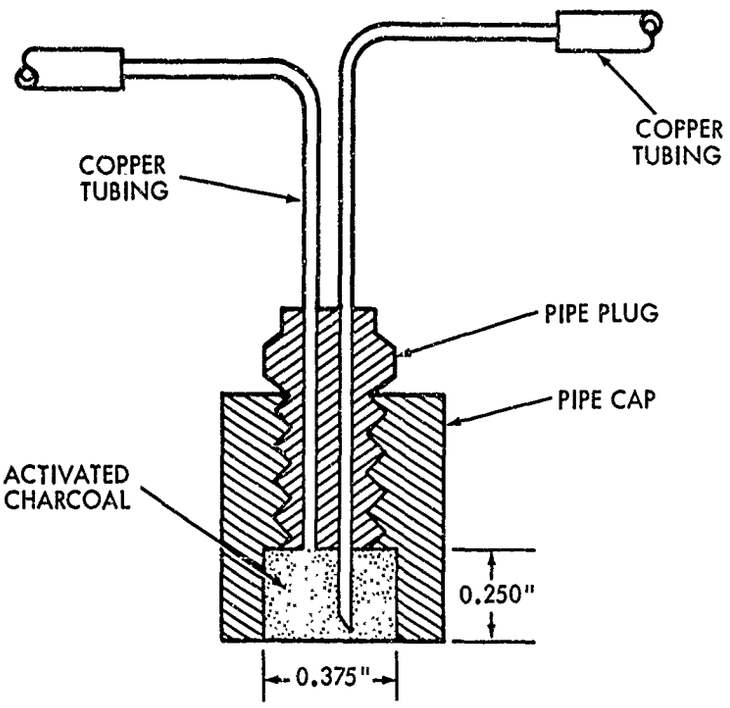

FIG. 2. GASEOUS FISSION PRODUCT COLLECTION TRAP.

soldered to the plug. One piece of copper tubing protruded about 3 mm into the cavity and the other tubing was even with the bottom of the plug. To the other ends of the copper tubing were silver-soldered short pieces of larger diameter copper tubing for convenient connection to rubber tubing. Copper tubing was used because it is more readily sealed than steel. Each collection trap was checked for leaks by pressurizing with helium at 20 psig.

Approximately 350 mg of activated charcoal, occupying a volume of 0.36 ml was introduced into the pipe plug. The plug was screwed tightly into the cap with the aid of teflon sealing tape. A drying tube containing silica gel was connected to each arm of the trap at least one day prior to experimental use. This was necessary to assure that the trap was sufficiently dry to prevent moisture from plugging it when cooled in liquid nitrogen.

In a typical experimental run the helium gas was allowed to flow continuously. The power for the vacuum pump and the Microflex timers was turned on. This closed the fission gas evacuation tubing at the point where it passed through the electro-hosecock and de-energized the solenoid which actuated the needle support.

The rabbit containing the uranyl stearate sample was driven by CO_2 gas pressure into the reactor for irradiation. The remote control starting switch initiating the Microflex timers (see Fig. 1) was closed manually at the midpoint of the 10 sec irradiation. The Microflex timers operated the equipment automatically for the next steps. The first Microflex timer introduced a precise 10 sec delay after closure of the switch. At the end of the 10 sec. delay period, the second Microflex timer was actuated. This energized the solenoid and drove the needles into the uranyl stearate package. Simultaneously, the electro-hosecock was opened, allowing helium to flow for 5 sec. At the end of 5 sec, the timers automatically closed the electro-hosecock and de-energized the solenoid circuit allowing the electro-hosecock to pinch shut the rubber tubing and simultaneously retracting the needles from the sample package. Finally, the two lengths of copper tubing on each gaseous collection trap were sealed with the aid of a hydraulic crimper. The irradiated samples were then allowed to decay for the desired period, removed from the rabbit and prepared for gamma spectral measurements.

Separate irradiations were utilized for measurements of the gaseous fission product fraction and of the solid fraction. Preparation of the solid fraction for gamma-ray measurements was simplified by not utilizing the gaseous fission product traps because of the tendency of the traps to plug. Several samples of the solid fractions were prepared for measurements at various times, and duplicate samples of the gaseous fission products were collected for measurements of that fraction.

DATA PROCESSING AND RESULTS

The processing of the experimental data was performed in a manner similar to that described previously (16,17). However, four different computers were used. A portion of the data was processed on a UNIVAC-1108 while the work was being done at the U. S. Naval Radiological Defense Laboratory.

When data processing was started at the Naval Ordnance Laboratory, an IBM-7090 was used. Later, the laboratory obtained a CDC-6400. During the period of changeover, Control Data Corporation made a CDC-6600 available. Consequently some modifications were made in the programs to accommodate them to each of the computers.

The gamma-ray spectra were obtained by unfolding the pulse-height distributions into 100 energy bins, identical with those utilized in the previous work (14, 15). The mid-bin energies and the widths of the energy bins are listed in Table 2.

The unfolded spectra from the fraction remaining after removal of the rare gases (i.e., the "solid" fraction) are given in Table 3. The results of the unfolding of the gamma-ray spectra of the species trapped on activated charcoal (i.e., the"gaseous" fraction) are contained in Table 4.

Figure 3 shows the pulse-height distributions of the two fractions at 1 hour post-fission. The distributions at 2 hours post-fission are given in Figure 4. Spectral-density histograms are given in Figures 5, 6, 7 and 8. These can be used to compare the results of this work with other results that vary in energy bin width.

DISCUSSION

The photon emission rates for the fractions obtained in this work are compared with the results for unfractionated fission products (17) in Table 5. The photon emission rate from the solid fraction is decreased about 9 to 28% over the 3 day time period with the smallest decrease at 3 days after fission. The members of several of the prominent mass chains removed by rare gas fractionation that exist at 3 days after fission are either stable, comparatively long-lived or pure β-ray emitters. Therefore it is suggested that the number of photons emitted by the solid fraction does not approach with time the number emitted by unfractionated fission products but will vary with increasing elapsed time after 3 days post-fission.

The sum of the photon emission rate from the gaseous fraction and that from the solid fraction is appreciably less than that from unfractionated fission products. The losses are ascribed to the deposition of fission products along the path of the gases from the

irradiated sample to the activated charcoal trap (see Figure 1). Radioactivity was observed all along the pathway. This deposition is believed to have resulted from the decay of the rare gas fission products to other elements which are not gaseous at room temperature. The atoms of these elements quickly adsorb onto surfaces. The solid fraction should give a more reliable quantitative measurement of the effects of fractionation than the gaseous fraction. However, qualitative indentification of constituents should be easier in the simpler spectra of the gaseous fractions.

The solid fractions obtained in the experimental work are considered to approximate reasonably well the radiochemical composition of fractionated close-in fallout. (Such fallout would most likely have a somewhat greater portion of the rare gas descendants than the solid fractions, but would also be depleted in mass chains which contained volatile members other than the rare gases.) Hence, the gamma-ray spectrum of the solid fraction should be similar to that emitted by close-in fallout material. Within limits, it is possible to make some comparisons of the results of this work with the calculations of Crocker (21). These are given in Table 6. Considering the assumptions involved, the agreement is good.

Examination of Figures 3 and 4 permits one to identify some prominent photopeaks in the pulse-height distributions of the gaseous fractions. The gamma-rays emitted in the decay of 92y probably contribute to the 0.45 and 0.55 MeV peaks, while the decay of ^{138}Xe would be expected to contribute to the 0.15, 0.25, and 0.45 MeV peaks. Contributions to the peaks at0.15, 0.45, 0.55, 1.02, 1.45, 2.25 and 2.65 MeV are expected from the decay of ^{138}Cs. Other contributors undoubtedly exist, but those mentioned above have the proper yields, genetic relationships, and half-lives to be prominent.

Table 2

Energy and Width of Energy Bins

Bin No.	Mid-bin Energy (MeV)	Width (MeV)	Bin No.	Mid-bin Energy (MeV)	Width (MeV)
1	0.07	0.01	51	1.470	0.05
2	0.08	0.01	52	1.520	0.05
3	0.09	0.01	53	1.570	0.05
4	0.10	0.01	54	1.620	0.05
5	0.11	0.01	55	1.670	0.05
6	0.12	0.01	56	1.720	0.05
7	0.13	0.01	57	1.770	0.05
8	0.14	0.01	58	1.820	0.05
9	0.15	0.01	59	1.870	0.05
10	0.16	0.01	60	1.925	0.06
11	0.17	0.01	61	1.985	0.06
12	0.18	0.01	62	2.045	0.06
13	0.19	0.01	63	2.105	0.06
14	0.20	0.01	64	2.165	0.06
15	0.215	0.02	65	2.225	0.06
16	0.235	0.02	66	2.285	0.06
17	0.255	0.02	67	2.345	0.06
18	0.275	0.02	68	2.405	0.06
19	0.295	0.02	69	2.465	0.06
20	0.315	0.02	70	2.525	0.06
21	0.335	0.02	71	2.585	0.06
22	0.355	0.02	72	2.645	0.06
23	0.375	0.02	73	2.705	0.06
24	0.395	0.02	74	2.770	0.07
25	0.420	0.03	75	2.840	0.07
26	0.450	0.03	76	2.910	0.07
27	0.480	0.03	77	2.980	0.07
28	0.510	0.03	78	3.050	0.07
29	0.540	0.03	79	3.120	0.07
30	0.570	0.03	80	3.190	0.07
31	0.600	0.03	81	3.265	0.08
32	0.630	0.03	82	3.345	0.08
33	0.665	0.04	83	3.425	0.08
34	0.705	0.04	84	3.505	0.08
35	0.745	0.04	85	3.585	0.08
36	0.785	0.04	86	3.665	0.08
37	0.825	0.04	87	3.750	0.09
38	0.865	0.04	88	3.840	0.09
39	0.905	0.04	89	3.930	0.09
40	0.945	0.04	90	4.020	0.09
41	0.985	0.04	91	4.115	0.10
42	1.025	0.04	92	4.215	0.10
43	1.070	0.05	93	4.315	0.10
44	1.120	0.05	94	4.415	0.10
45	1.170	0.05	95	4.515	0.10
46	1.220	0.05	96	4.615	0.10
47	1.270	0.05	97	4.715	0.10
48	1.320	0.05	98	4.820	0.11
49	1.370	0.05	99	4.930	0.11
50	1.420	0.05	100	5.040	0.11

TABLE 3

GAMMA-RAY SPECTRA OF SOLID FRACTION OF U-235 FISSION PRODUCTS
[PHOTONS/(FISSION SEC)]

ENERGY BIN NO.	TIME AFTER FISSION								
	¼ H	½ H	1 H	2 H	5 H	10 H	24 H	48 H	72 H
1	(5)49	(5)26	(6)39	(6)43	(7)69	(7)42	(8)80	(7)24	(7)13
2	(5)65	(5)37	(5)21	(6)51	(7)38	(7)18	(8)43	(7)17	(8)80
3	(5)37	(6)89	(6)57	(6)24	(7)32	(7)24	(7)11	(7)14	(7)10
4	(5)24	(5)10	(6)58	(6)20	(7)16	(7)12	(8)67	(8)82	(8)48
5	(5)18	(6)53	(6)27	(6)31	(7)29	(7)14	(7)12	(7)10	(8)60
6	(5)16	(6)74	(6)28	(6)15	(7)34	(7)24	(7)24	(7)19	(7)21
7	(5)18	(5)11	(6)61	(6)36	(7)65	(7)59	(7)67	(7)70	(7)46
8	(5)29	(5)22	(5)15	(6)73	(7)86	(7)66	(7)60	(7)51	(7)40
9	(5)42	(5)21	(5)17	(6)37	(7)57	(7)26	(7)18	(7)12	(7)16
10	(5)37	(5)19	(5)12	(6)34	(7)64	(7)11	(8)66	(8)46	(8)27
11	(5)61	(5)40	(5)15	(6)32	(7)51	(8)88	(8)57	(8)49	(8)24
12	(4)11	(5)56	(5)19	(6)38	(7)30	(8)87	(8)64	(8)71	(8)46
13	(4)10	(5)51	(5)24	(6)57	(7)35	(8)87	(8)42	(8)35	(8)40
14	(5)80	(5)50	(5)22	(6)67	(7)69	(7)22	(8)80	(8)60	(8)37
15	(4)10	(5)41	(5)23	(6)67	(6)16	(7)94	(7)52	(7)58	(7)44
16	(5)61	(5)23	(6)82	(6)31	(6)28	(6)26	(7)44	(7)71	(7)30
17	(4)12	(5)58	(5)19	(6)68	(6)26	(6)23	(7)19	(7)32	(8)78
18	(4)12	(5)51	(5)26	(5)10	(6)19	(6)11	(7)57	(7)42	(7)21
19	(4)23	(4)18	(5)70	(5)12	(6)18	(6)12	(7)70	(7)48	(7)29
20	(4)29	(4)16	(5)66	(5)10	(6)10	(7)35	(7)18	(7)12	(7)10
21	(5)85	(5)40	(5)18	(6)39	(7)62	(7)23	(7)15	(7)11	(8)90
22	(4)10	(5)51	(5)16	(6)22	(7)49	(7)38	(7)25	(7)23	(7)16
23	(5)49	(5)19	(6)86	(6)21	(7)36	(7)16	(7)10	(8)83	(8)85
24	(5)47	(5)18	(6)78	(6)40	(7)71	(7)14	(8)52	(8)27	(8)21
25	(4)13	(5)58	(5)29	(5)13	(6)21	(7)48	(8)75	(8)34	(10)82

TABLE 3 (Continued)

ENERGY BIN NO.	TIME AFTER FISSION								
	¼ H	½ H	1 H	2 H	5 H	10 H	24 H	48 H	72 H
26	(4)15	(4)10	(5)57	(5)17	(6)15	(7)50	(8)82	(8)58	(8)61
27	(4)10	(5)48	(5)23	(6)55	(6)11	(7)49	(7)25	(7)15	(7)12
28	(5)65	(5)33	(5)13	(6)74	(6)35	(6)25	(6)15	(7)83	(7)44
29	(5)60	(5)44	(5)31	(5)16	(6)77	(6)54	(6)23	(6)10	(7)44
30	(4)16	(5)72	(5)27	(5)10	(6)39	(6)19	(7)71	(7)10	(8)60
31	(4)17	(5)61	(5)21	(6)82	(6)16	(7)47	(7)18	(8)69	(8)49
32	(4)12	(5)78	(5)50	(5)28	(6)54	(6)15	(7)88	(7)48	(7)27
33	(5)92	(5)60	(5)36	(5)18	(5)10	(6)60	(6)30	(6)16	(7)82
34	(4)15	(5)45	(5)17	(6)67	(6)33	(6)21	(6)14	(7)56	(7)42
35	(4)16	(5)89	(5)55	(5)26	(6)72	(6)48	(6)24	(6)12	(7)55
36	(4)16	(5)76	(5)37	(5)13	(6)48	(6)28	(6)16	(7)81	(7)59
37	(4)16	(5)62	(5)40	(5)28	(6)33	(7)56	(7)23	(7)11	(7)11
38	(4)25	(4)14	(4)11	(5)76	(5)12	(6)11	(7)31	(7)14	(7)10
39	(4)28	(4)15	(5)76	(5)27	(6)69	(6)10	(7)24	(7)10	(8)56
40	(4)23	(4)12	(5)46	(5)13	(6)40	(6)14	(7)31	(7)12	(8)85
41	(4)12	(5)46	(5)17	(6)66	(6)22	(6)10	(7)33	(7)13	(8)83
42	(4)12	(5)58	(5)20	(6)90	(6)28	(6)13	(7)45	(7)11	(8)50
43	(4)11	(5)54	(5)24	(5)13	(6)36	(6)13	(7)42	(8)84	(8)38
44	(4)10	(5)43	(5)25	(5)13	(6)40	(6)14	(7)36	(7)10	(8)51
45	(4)11	(5)48	(5)17	(6)58	(6)30	(6)13	(7)35	(8)81	(8)52
46	(4)12	(5)48	(5)14	(6)43	(6)14	(7)52	(7)20	(8)54	(8)30
47	(4)10	(5)43	(5)12	(6)45	(6)30	(6)16	(7)38	(8)79	(8)31
48	(5)83	(5)36	(5)13	(6)43	(6)19	(7)95	(7)25	(8)73	(8)41
49	(5)90	(5)52	(5)36	(5)23	(6)61	(6)16	(7)16	(8)67	(8)44
50	(5)75	(5)50	(5)24	(5)13	(6)86	(6)27	(7)21	(8)66	(8)40

TABLE 3 (Continued)

ENERGY BIN NO.	TIME AFTER FISSION								
	¼ H	½ H	1 H	2 H	5 H	10 H	24 H	48 H	72 H
51	(5)48	(5)27	(5)11	(6)33	(6)17	(7)72	(7)16	(8)33	(8)22
52	(5)45	(5)25	(5)12	(6)32	(7)71	(7)29	(8)91	(8)24	(8)17
53	(5)42	(5)23	(5)10	(6)35	(7)64	(7)19	(8)76	(8)60	(8)62
54	(5)39	(5)19	(6)84	(6)33	(7)71	(7)21	(7)11	(8)95	(7)11
55	(5)54	(5)23	(5)10	(6)38	(6)11	(7)43	(7)14	(8)48	(8)47
56	(5)51	(5)23	(5)10	(6)45	(6)15	(7)68	(7)14	(8)24	(8)11
57	(5)33	(5)16	(6)79	(6)44	(6)15	(7)56	(7)13	(8)17	(9)41
58	(5)30	(5)13	(6)68	(6)34	(6)12	(7)41	(7)10	(8)16	(9)48
59	(5)32	(5)14	(6)74	(6)29	(6)10	(7)30	(8)73	(8)16	(9)90
60	(5)36	(5)18	(6)70	(6)24	(7)81	(7)23	(8)61	(8)17	(8)11
61	(5)39	(5)18	(6)67	(6)21	(7)66	(7)18	(8)48	(8)15	(9)92
62	(5)34	(5)15	(6)64	(6)22	(7)63	(7)14	(8)33	(8)13	(8)11
63	(5)32	(5)13	(6)55	(6)20	(7)62	(7)13	(8)26	(8)12	(8)11
64	(5)41	(5)18	(6)57	(6)18	(7)55	(7)11	(8)23	(9)86	(9)65
65	(5)31	(5)17	(6)45	(6)14	(7)42	(8)95	(8)22	(9)67	(9)39
66	(5)19	(6)89	(6)34	(6)12	(7)32	(8)83	(8)21	(9)63	(9)49
67	(5)17	(6)73	(6)44	(6)21	(7)40	(8)88	(8)19	(9)62	(9)52
68	(5)16	(6)77	(6)57	(6)30	(7)68	(7)10	(8)18	(9)59	(9)55
69	(5)16	(6)76	(6)51	(6)24	(7)76	(7)10	(8)16	(9)54	(9)51
70	(5)19	(6)88	(6)44	(6)19	(7)59	(8)86	(8)13	(9)49	(9)48
71	(5)24	(5)10	(6)36	(6)14	(7)46	(8)64	(8)11	(9)42	(9)45
72	(5)23	(6)90	(6)26	(7)88	(7)32	(8)50	(9)92	(9)35	(9)36
73	(5)15	(6)63	(6)17	(7)59	(7)21	(8)43	(9)87	(9)31	(9)19
74	(6)94	(6)37	(6)13	(7)50	(7)16	(8)40	(9)85	(9)29	(10)39
75	(6)68	(6)27	(6)13	(7)56	(7)15	(8)36	(9)78	(9)26	0

TABLE 3 (Continued)

ENERGY BIN NO.	TIME AFTER FISSION								
	¼ H	½ H	1 H	2 H	5 H	10 H	24 H	48 H	72 H
76	(6)51	(6)25	(6)14	(7)67	(7)17	(8)31	(9)63	(9)23	(9)11
77	(6)51	(6)26	(6)15	(7)74	(7)20	(8)28	(9)54	(9)21	(9)21
78	(6)56	(6)27	(6)13	(7)57	(7)20	(8)25	(9)47	(9)18	(9)24
79	(6)60	(6)26	(6)10	(7)37	(7)14	(8)21	(9)43	(9)17	(9)23
80	(6)58	(6)23	(7)81	(7)29	(7)10	(8)19	(9)41	(9)17	(9)16
81	(6)54	(6)20	(7)72	(7)26	(8)86	(8)17	(9)38	(9)17	(10)37
82	(6)46	(6)17	(7)66	(7)25	(8)81	(8)16	(9)34	(9)17	(12)90
83	(6)46	(6)16	(7)62	(7)23	(8)75	(8)14	(9)32	(9)17	(10)85
84	(6)60	(6)21	(7)62	(7)21	(8)69	(8)13	(9)31	(9)16	(10)90
85	(6)62	(6)23	(7)57	(7)20	(8)65	(8)12	(9)30	(9)16	(10)36
86	(6)40	(6)16	(7)47	(7)17	(8)61	(8)11	(9)28	(9)15	0
87	(6)25	(6)11	(7)38	(7)14	(8)53	(8)10	(9)26	(9)15	0
88	(6)22	(7)91	(7)35	(7)12	(8)44	(8)10	(9)24	(9)15	0
89	(6)24	(7)93	(7)34	(7)11	(8)38	(9)90	(9)24	(9)15	(10)65
90	(6)27	(6)10	(7)30	(7)10	(8)35	(9)86	(9)24	(9)16	(9)14
91	(6)25	(7)89	(7)26	(8)91	(8)33	(9)86	(9)25	(9)16	(9)11
92	(6)19	(7)71	(7)23	(8)85	(8)31	(9)85	(9)25	(9)16	(10)92
93	(6)15	(7)59	(7)22	(8)82	(8)30	(9)83	(9)25	(9)16	(9)12
94	(6)13	(7)53	(7)22	(8)80	(8)29	(9)83	(9)27	(9)17	(9)10
95	(6)12	(7)50	(7)21	(8)79	(8)28	(9)86	(9)28	(9)17	(10)53
96	(6)13	(7)51	(7)21	(8)83	(8)30	(8)10	(9)31	(9)19	(10)35
97	(6)13	(7)54	(7)23	(8)90	(8)33	(8)11	(9)35	(9)21	0
98	(6)14	(7)57	(7)25	(7)10	(8)36	(8)12	(9)40	(9)23	0
99	(6)15	(7)63	(7)27	(7)11	(8)40	(8)14	(9)47	(9)27	0
100	(6)19	(7)81	(7)36	(7)14	(8)51	(8)18	(9)61	(9)34	0

TABLE 4

GAMMA-RAY SPECTRA OF GASEOUS FRACTION OF ^{235}U FISSION PRODUCTS
[PHOTONS/(FISSION SEC)]

ENERGY BIN NO.	TIME AFTER FISSION 1 H	2 H	ENERGY BIN NO.	TIME AFTER FISSION 1 H	2 H
1	(7)33	(8)94	26	(5)12	(6)31
2	(8)24	(8)18	27	(6)86	(6)31
3	(11)25	(9)55	28	(6)15	(7)70
4	(12)62	(8)16	29	(6)41	(6)12
5	(9)34	(8)29	30	(6)29	(6)12
6	(7)28	(8)94	31	(7)37	(7)23
7	(7)86	(7)28	32	(7)48	(8)93
8	(6)13	(7)27	33	(6)19	(7)22
9	(6)17	(7)59	34	(7)71	(7)24
10	(6)17	(7)80	35	(7)37	(7)23
11	(6)14	(7)62	36	(7)92	(7)32
12	(6)11	(7)54	37	(7)94	(7)33
13	(6)12	(7)62	38	(6)21	(7)79
14	(6)14	(7)59	39	(6)28	(6)10
15	(6)19	(7)64	40	(6)14	(7)46
16	(6)37	(7)76	41	(6)41	(7)70
17	(6)56	(7)50	42	(5)15	(6)33
18	(6)20	(7)47	43	(6)81	(6)24
19	(7)76	(7)16	44	(6)10	(7)19
20	(7)70	(7)18	45	(7)73	(7)41
21	(7)72	(7)36	46	(6)31	(7)38
22	(6)10	(7)14	47	(6)48	(7)28
23	(6)12	(7)34	48	(6)19	(7)31
24	(6)35	(7)94	49	(6)21	(7)54
25	(6)56	(6)11	50	(5)17	(6)32

TABLE 4 (Continued)

ENERGY BIN NO.	TIME AFTER FISSION 1 H	2 H	ENERGY BIN NO.	TIME AFTER FISSION 1 H	2 H
51	(5)27	(6)92	76	(7)19	(8)56
52	(6)48	(6)40	77	(7)18	(8)48
53	(6)11	(7)44	78	(7)20	(8)53
54	(7)30	(8)78	79	(7)25	(8)63
55	(7)20	(8)45	80	(7)29	(8)71
56	(7)53	(8)55	81	(7)29	(8)75
57	(6)18	(7)12	82	(7)25	(8)75
58	(6)24	(7)34	83	(7)22	(8)74
59	(6)15	(7)65	84	(7)18	(8)61
60	(6)10	(7)53	85	(7)14	(8)45
61	(6)18	(7)27	86	(7)12	(8)38
62	(6)31	(7)36	87	(7)12	(8)39
63	(6)19	(7)45	88	(7)13	(8)44
64	(6)25	(7)47	89	(7)13	(8)44
65	(6)67	(6)14	90	(7)12	(8)39
66	(6)57	(6)24	91	(7)10	(8)34
67	(6)16	(7)84	92	(8)84	(8)30
68	(7)78	(7)51	93	(8)76	(8)27
69	(7)70	(7)60	94	(8)70	(8)25
70	(7)81	(7)38	95	(8)67	(8)22
71	(6)15	(7)25	96	(8)72	(8)22
72	(6)31	(7)47	97	(8)82	(8)25
73	(6)28	(6)10	98	(8)90	(8)29
74	(6)10	(7)56	99	(8)92	(8)32
75	(7)30	(7)13	100	(7)11	(8)36

TABLE 5

PHOTON EMISSION RATES OF PRODUCTS
OF THERMAL-NEUTRON FISSION OF ^{235}U

Time After Fission (hr)	Photons/Fission-Sec			Fraction of Gross Fission Products	
	Gaseous Fraction	Solid Fraction	Gross[a] Fission Products	Gaseous[b] Fraction	Solid Fraction
0.25	---	6.2×10^{-4}	8.1×10^{-4}	--	0.77
0.5	---	3.1×10^{-4}	4.3×10^{-4}	--	0.72
1	2.2×10^{-5}	1.5×10^{-4}	2.0×10^{-4}	0.11	0.75
2	6.2×10^{-6}	5.9×10^{-5}	7.9×10^{-5}	0.08	0.75
5	---	1.6×10^{-5}	2.0×10^{-5}	--	0.80
10	---	6.5×10^{-6}	8.1×10^{-6}	--	0.80
24	---	2.6×10^{-6}	3.6×10^{-6}	--	0.72
48	---	1.4×10^{-6}	1.6×10^{-6}	--	0.88
72	---	8.6×10^{-7}	9.4×10^{-7}	--	0.91

[a]From unfractionated products of thermal-neutron fission given in reference 17.

[b]These values are low due to loss of a portion of the activity in the gas fraction that was deposited in the needle and tubing leading to the trap.

TABLE 6

PHOTON EMISSION RATES OF "FRACTIONATED FALLOUT"
PHOTONS/FISSION - SEC

Time After Fission (hr)	Solid Fraction	Calculated (Moderate Loss of Chains)	Experimental Calculated
1	1.5×10^{-4}	1.0×10^{-4}	1.5
2	5.9×10^{-4}	4.1×10^{-5}	1.2
5	1.6×10^{-5}	1.2×10^{-5}	1.3
24	2.6×10^{-6}	2.9×10^{-6}	0.90
48	1.4×10^{-6}	1.5×10^{-6}	0.93
72	8.6×10^{-7}	9.7×10^{-7}	0.89

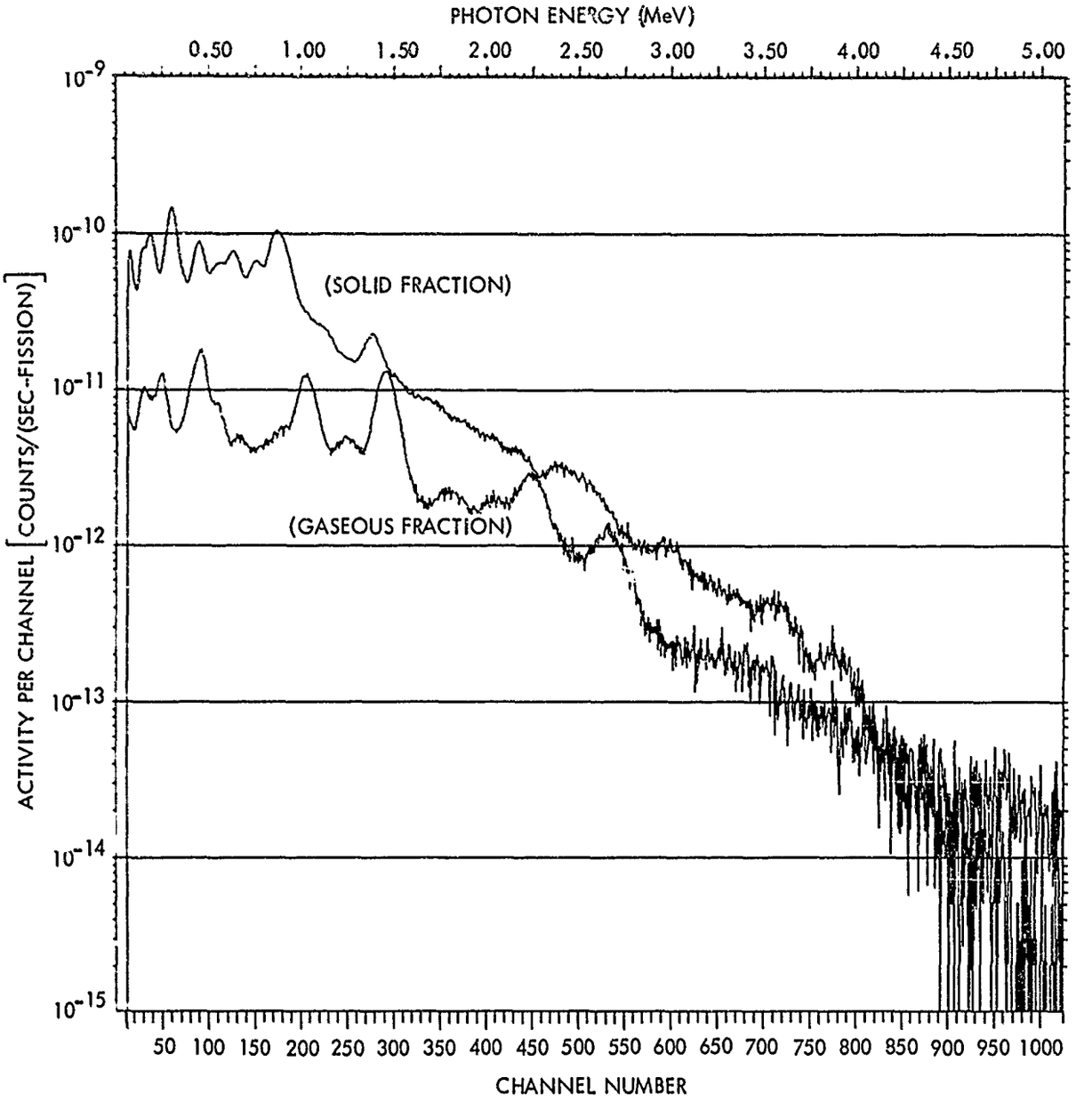

FIG. 3 PULSE-HEIGHT DISTRIBUTIONS OF FRACTIONATED PRODUCTS OF THERMAL-NEUTRON FISSION OF ^{235}U AT 1 HOUR AFTER FISSION.

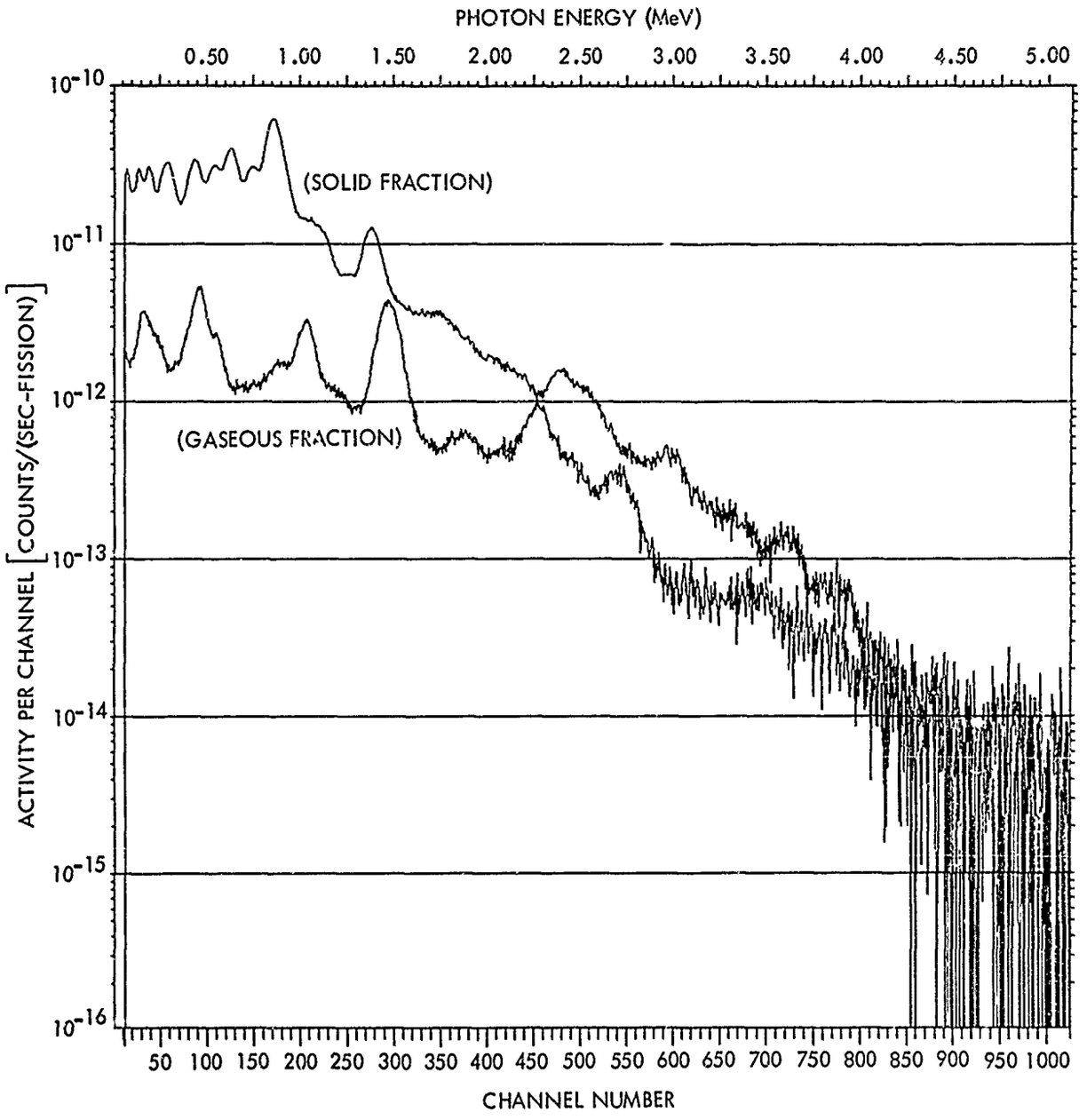

FIG. 4 PULSE-HEIGHT DISTRIBUTIONS OF FRACTIONATED PRODUCTS OF THERMAL-NEUTRON FISSION OF ^{235}U AT 2 HOURS AFTER FISSION.

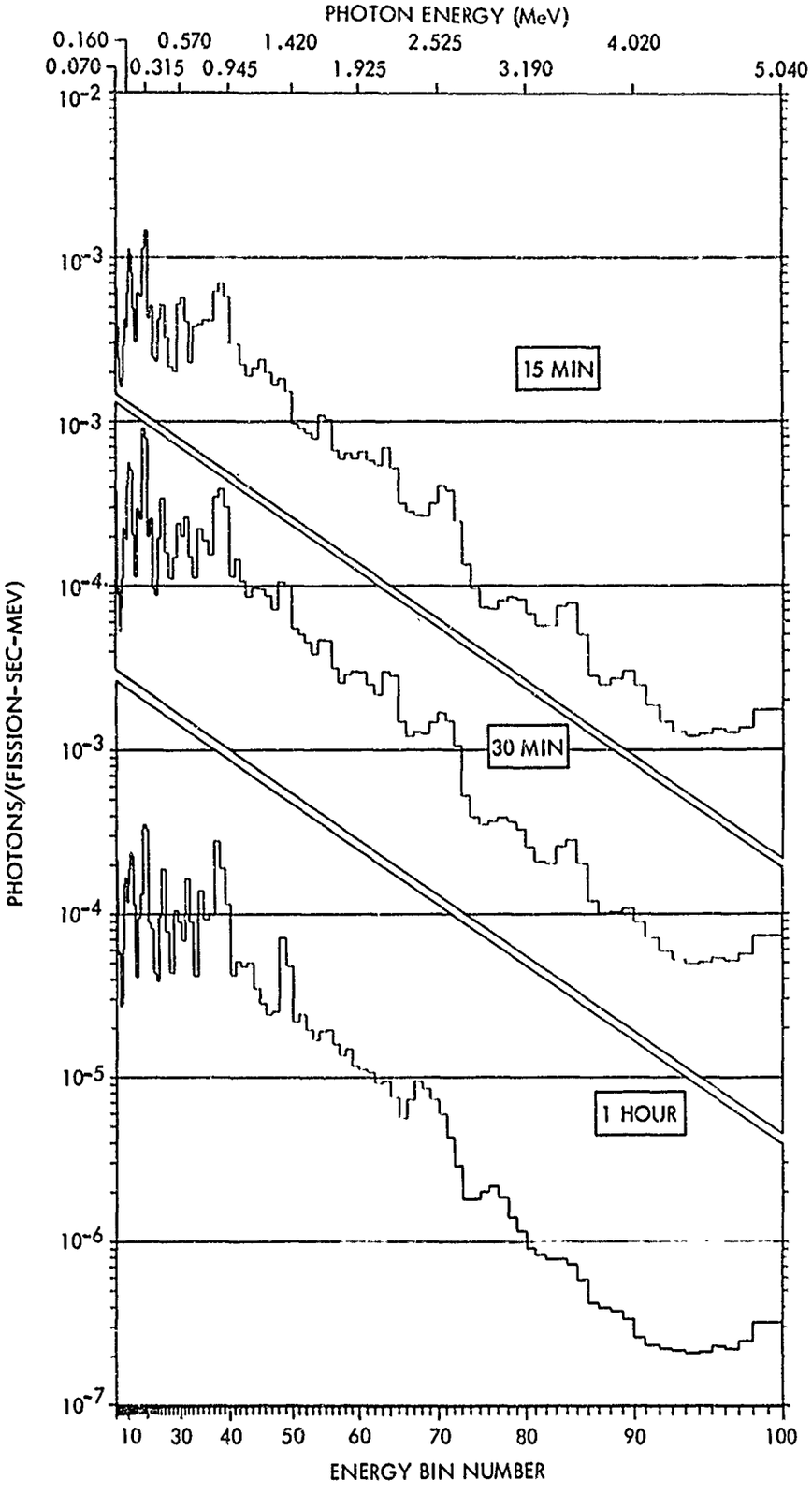

FIG. 5 GAMMA-RAY SPECTRAL-DENSITY HISTOGRAMS OF THE SOLID FRACTIONS FROM PRODUCTS OF THERMAL-NEUTRON FISSION OF ^{235}U AT SELECTED TIMES AFTER FISSION.

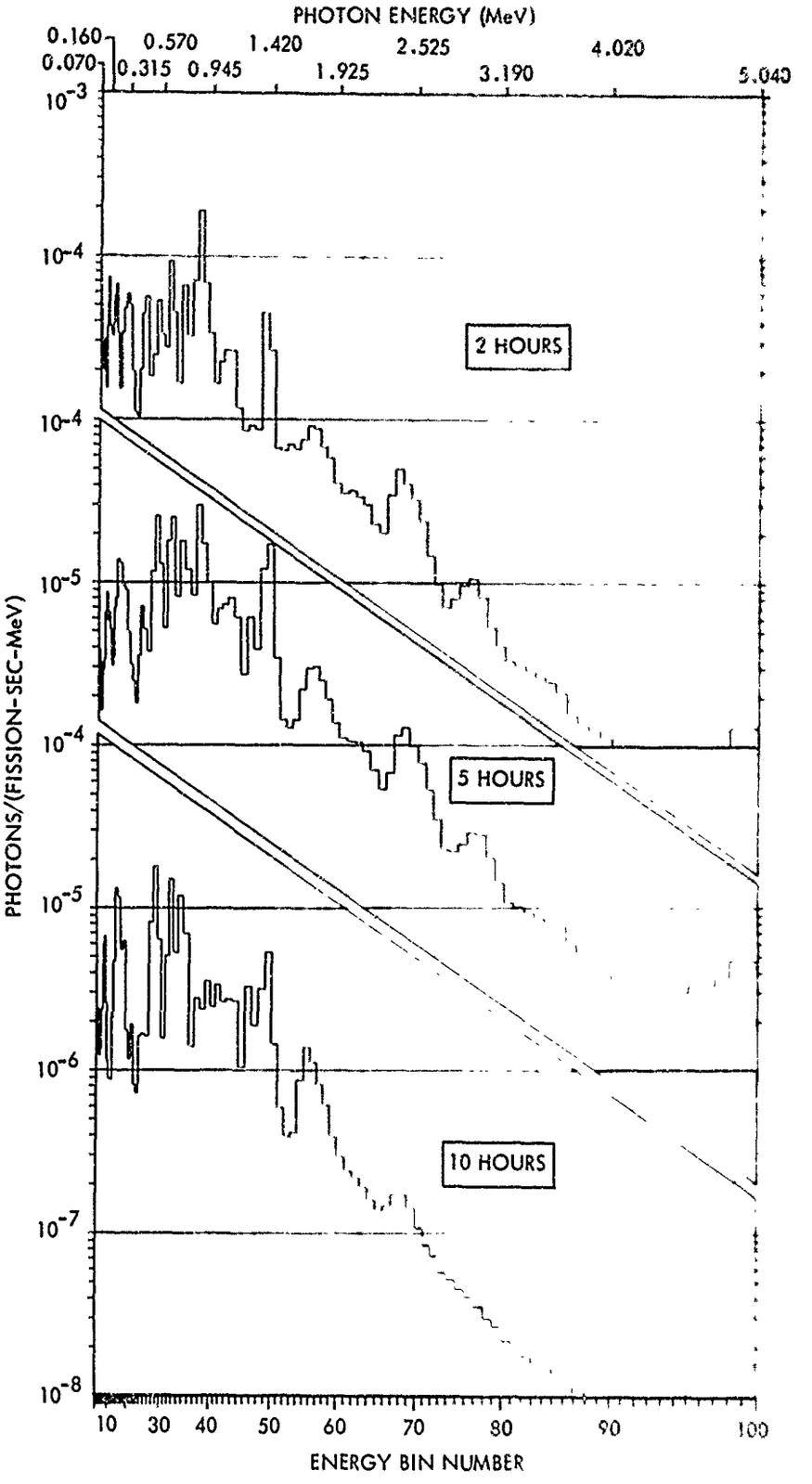

FIG. 6. GAMMA-RAY SPECTRAL-DENSITY HISTOGRAMS OF THE SOLID FRACTIONS FROM PRODUCTS OF THERMAL-NEUTRON FISSION OF ^{235}U AT SELECTED TIMES AFTER FISSION.

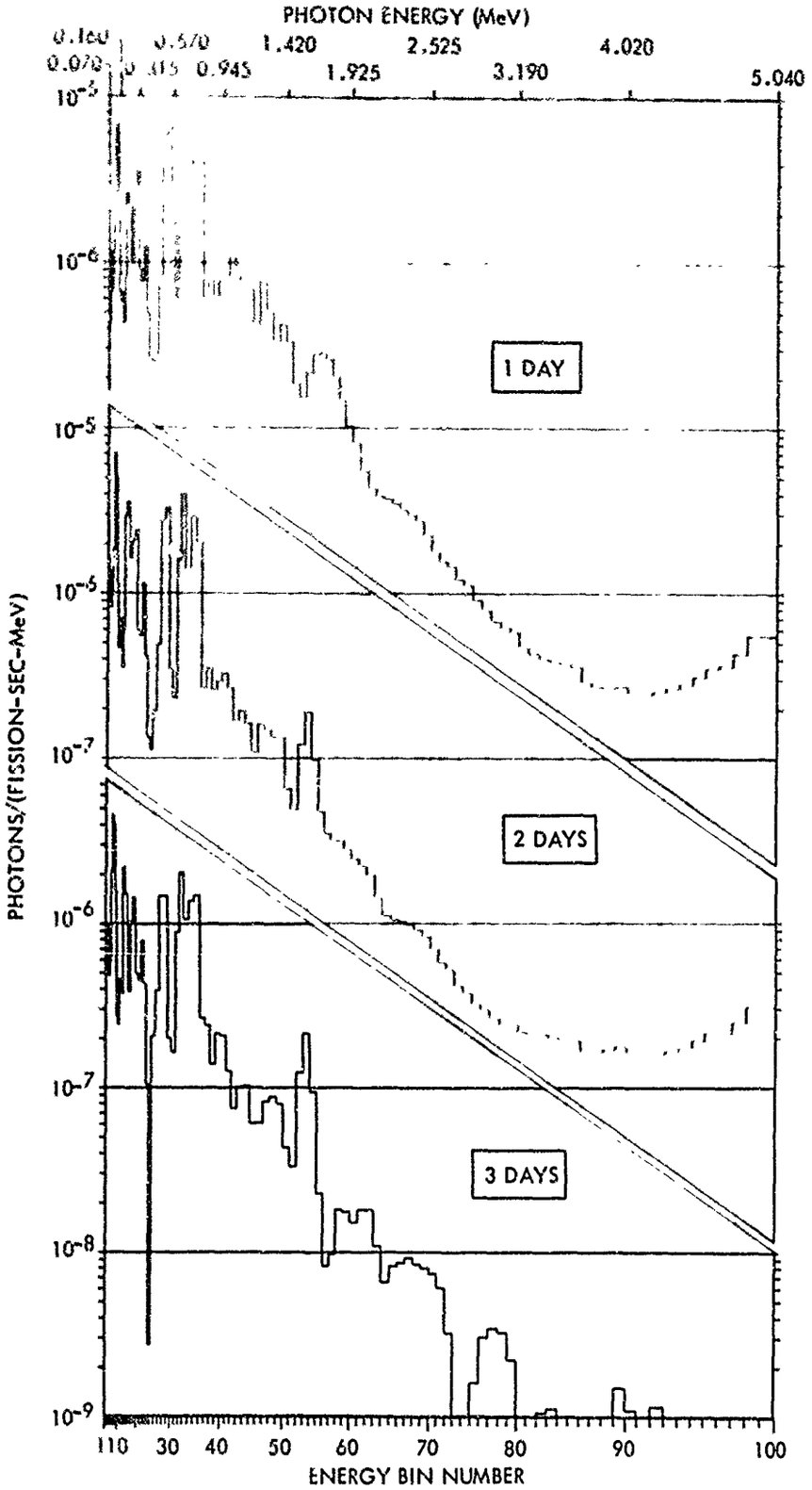

FIG. 7 GAMMA-RAY SPECTRAL-DENSITY HISTOGRAMS OF THE SOLID FRACTIONS FROM PRODUCTS OF THERMAL-NEUTRON FISSION OF ^{235}U AT SELECTED TIMES AFTER FISSION.

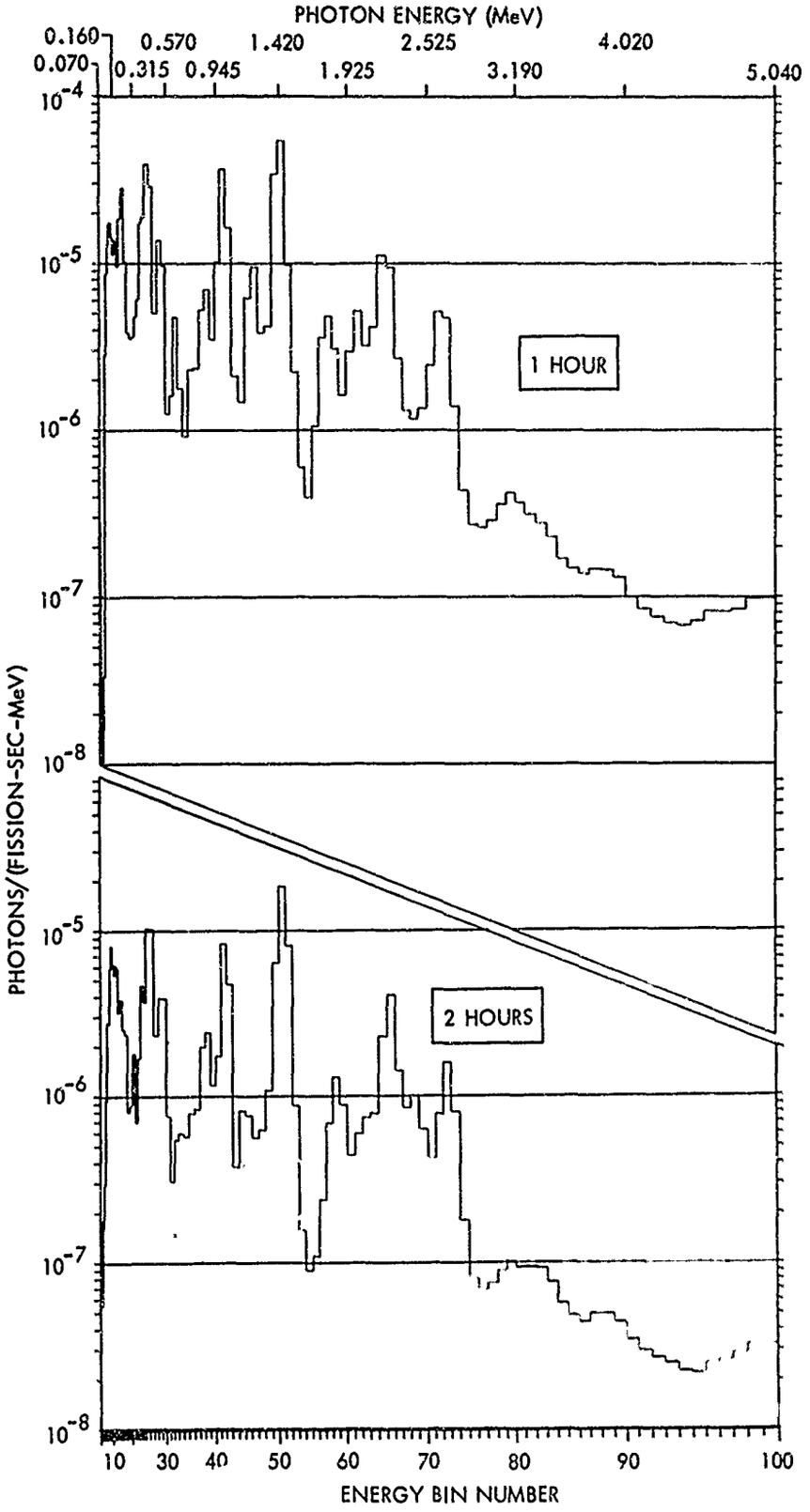

FIG. 8 GAMMA-RAY SPECTRAL-DENSITY HISTOGRAMS OF THE GASEOUS FRACTIONS FROM THERMAL-NEUTRON FISSION OF ^{235}U AT 1 HOUR AND 2 HOURS AFTER FISSION.

REFERENCES

1. Ballou, N. E., and Bunney, L. R., "Nature and Distribution of Residual Contamination. II", WT-397, 30 June 1952.

2. Whitcher, S. L., Bunney, L. R., Soule, R. R., and daRoza, R. A., "Fallout Measurements by Aircraft and Rocket Sampling", WT-1625, 29 September 1961.

3. Freiling, E. C., Bunney, L. R., and Kawahara, F. K., "Physicochemical and Radiochemical Analysis", POR-2216, 15 October 1964.

4. Freiling, E. C., "Radionuclide Fractionation in Bomb Debris", Science 133, 1991 (1961).

5. Freiling, E. C. and Rainey, S. C., "Fractionation II. On Defining the Surface Density of Contamination", USNRDL-TR-631, January 1963.

6. Freiling, E. C., "Fractionation III. Estimation of Degree of Fractionation and Radionuclide Partition for Nuclear Debris", USNRDL-TR-680, 12 September 1963.

7. Adams, C. E., Wallace, N. R., and Popoff, I. G., "The Nature of Individual Radioactive Particles I. Surface and Underground ABD Particles from Operation JANGLE", USNRDL-374, 1952.

8. Adams, C. E., Farlow, N. H., and Schell, W. R., "The Compositions, Structures and Origin of Radioactive Fallout Particles", Geochimica Et Cosmochimica Acta 18, 42 (1960.

9. Norman, J. H. and Winchell, P., "Cloud Chemistry of Fallout Formation", GA-7051, 1966.

10. Norman, J. H. and Winchell, P., "Physical, Chemical, and Radiological Properties of Fallout", GA-9741, 24 July 1970.

11. "Radioactive Fallout from Nuclear Weapons Tests", Edited by A. W. Klement, U. S. Atomic Energy Commission, November 1965.

12. "Radionuclides in the Environment", Edited by E. C. Freiling, Advances in Chemistry Series 93, American Chemical Society, Washington, D. C. 1970

13. Crocker, G. R., "Radiation Properties of Fractionated Fallout, Predictions of Activities, Exposure Rates, and Gamma Spectra for Selected Situations" NRDL-TR-68-134, 27 June 1968.

14. Bunney, L. R., and Sam, D., "Gamma-Ray Spectra of the Products of Fast Neutron Fission of U^{235} and U^{238} at Selected Times After Fission", Nucl. Sci. Eng. 29, 432 (1967).

15. Bunney, L. R., and Sam, D., "Exposure Rates from the Products of Fast Neutron Fission of ^{235}U and ^{238}U at Selected Temes After Fission" Health Phys. 13, 1033 (1967).

16. Sam, D., Bunney, L. R., and Heater, D. C., "Gamma-Ray Pulse-Height Spectra: Formation of a Response Matrix for Interative Unfolding", Nucl. Instr. Methods 64, 148 (1968).

17. Bunney, L. R., and Sam, D., "Gamma-Ray Spectra of The Products of Thermal Neutron Fission of ^{235}U at Selected Times After Fission", Nucl. Sci. Eng. 39, 81 (1970).

18. Sam, D. and Bunney, L. R., "Exposure Rates from the Products of Thermal-Neutron Fission of ^{235}U at Selected Times after Fission", NRDL-TR-69-60, 9 June 1969.

19. Wahl, A. C., and Daniels, W. R., "Nuclear-Charge Distribution in Fission: Cumulative Yields of Short-Lived Krypton and Kenon Isotopes from Thermal-Neutron Fission of ^{235}U", J. Inorg. and Nucl. Chem. 6, 278 (1958).

20. Wahl, A. C., "Emanating Power of Barium Stearate for 3.9-Second Actinon (^{219}Ru)", J. Inorg. and Nucl. Chem. 6, 263 (1958).

21. Crocker, G. R., "Radiation Properties of Fractionated Fallout. Predictions of Activities, Exposure Rates and Gamma Spectra for Selected Situations", NRDL-TR-68-134, 27 June 1968.